INVESTIGATION OF LASER DYNAMICS, MODULATION AND CONTROL BY MEANS OF INTRA-CAVITY TIME VARYING PERTURBATION

under the direction of

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Semiannual Status Report

(Report No. 20)

for

NASA Grant NGL-05-020-103

National Aeronautics and Space Administration

Washington, D.C.

for the period

1 August 1973 – 31 January 1974

M. L. Report No. 2259

January 1974

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NASA Grant NGL-05-020-103

for the period

1 August 1973 – 31 January 1974

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I. INTRODUCTION

Work continues on a number of projects aimed at the generation of tunable visible, infrared, and ultraviolet light, and on the control of this light by means of novel mode-locking and modulation techniques. During this period the following projects have been active: (1) studies of transient mode-locking of the Nd:YAG laser and the application of short optical pulses; (2) experimental investigations of the Na-Xe excimer laser system; (3) further development of techniques for vacuum ultraviolet holography; and (4) theoretical and initial experimental studies of a new device which should prove very useful for both infrared up-conversion and generation of tunable UV radiation — a two-photon resonantly pumped frequency converter. Progress on each of these projects is summarized in the following sections.

II. SUMMARY OF PROGRESS

A. Transient Mode-Locking in the Pre-Lasing Mode

(D. W. Phillion, D. J. Kuizenga, and A. E. Siegman)

In previous semiannual reports, the theoretical basis of transient mode-locking has been advanced and supporting experimental data given. A preprint of the paper describing the theory and measurements was included as Appendix A of the last status report and has since been published in Optics Communications.[1]

During this period the behavior of a Q-switched and mode-locked laser has been studied in the pre-lasing mode, in which there is a period of cw oscillation prior to each Q-switched pulse. The mode-locked pulses evolve to the steady-state during the interval of cw lasing and are amplified unchanged in shape when the Q-switch is turned off, giving an additional peak power enhancement of 10^4. According to the theoretical model of transient mode-locking described previously, each mode-locked pulse in the Q-switched burst will have a width

$$\tau_p = \frac{\sqrt{2 \ln 2}}{\pi} \left(\frac{g}{\theta^2}\right)^{1/4} \left(\frac{1}{\Delta f_a f_m}\right)^{1/2} , \qquad (1)$$

[1] D. J. Kuizenga, D. W. Phillion, T. Lund, and A. E. Siegman, "Simultaneous Q-Switching and Mode-Locking in the CW Nd:YAG Laser," Optics Comm. 9, 221 (November 1973).

in which g is the round-trip amplitude gain at atomic line center during the build-up of the Q-switched pulse, Δf_a is the full width at half maximum of the Lorentzian line, and f_m is the RF driving frequency to the amplitude modulator, here one half the axial mode spacing Δf_{axial}. The single pass amplitude transmission through the acousto-optic modulator is $\cos(\theta \sin \omega_m t)$, assuming perfect Bragg diffraction.

The results summarized by Fig. 1 demonstrate a good absolute agreement with theoretical predictions and an even better adherence to the power law relation $\tau \propto \theta^{-1/2}$ or $\tau \propto P_{RF}^{-1/4}$, in which P_{RF} is the RF driving power to the modulator. The only parameter not sufficiently well-known which could account for the 15% absolute discrepancy with theory is the linewidth Δf_a, which has a strong temperature dependence.

To satisfy the group velocity condition requires that the period T between successive transmission maxima of the amplitude modulator be equal to the cavity round-trip time $2L\,\overline{v_g^{-1}}$, in which L is the length of the Fabry-Perot laser cavity and $\overline{v_g^{-1}}$ is the mean reciprocal group velocity. The population inversion of the $^4F_{3/2} \rightarrow {}^4I_{11/2}$ lasing transition (1.064μ) contributes to the dispersion $\partial k/\partial \omega$ and thus the cavity length required to meet the group velocity condition depends on gain. The modulation frequency f_m can also be varied to satisfy this condition. The population inversion will shift this frequency by an amount

$$\delta f_m = \frac{-2f_m^2 g}{\pi \Delta f_a} \quad . \tag{2}$$

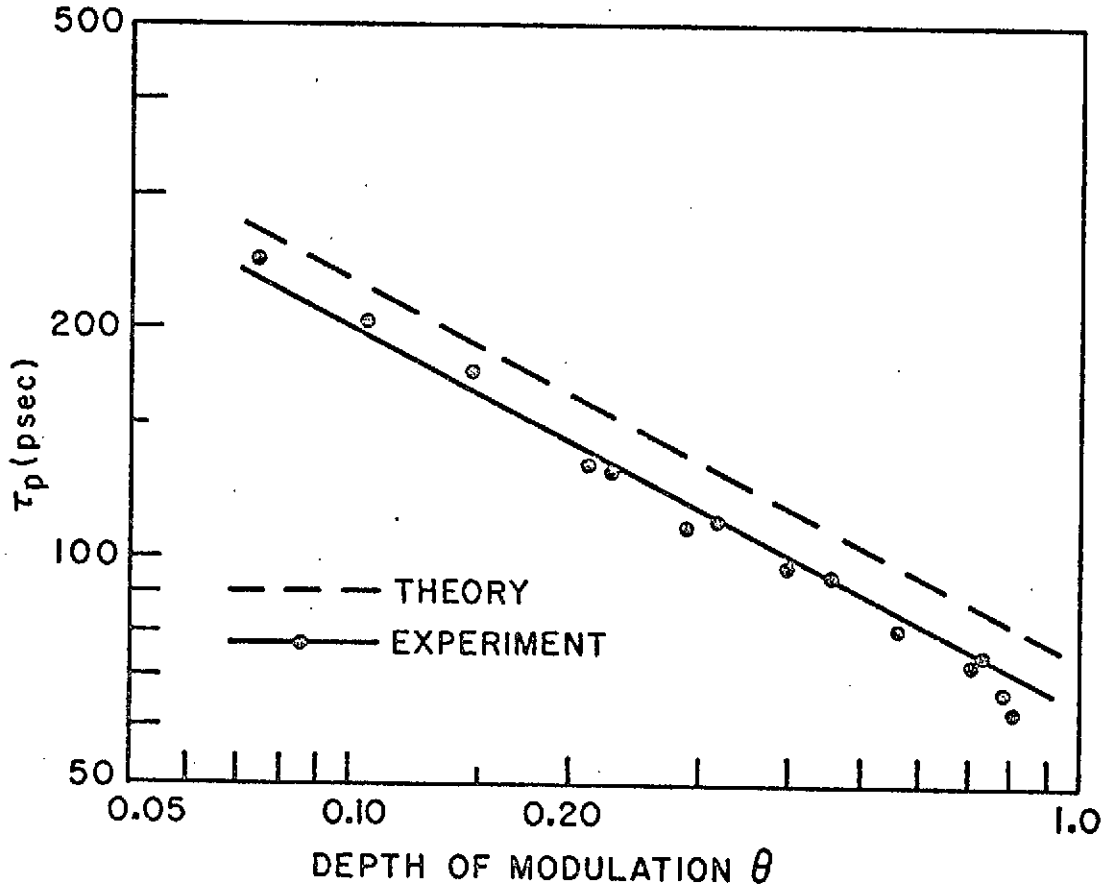

FIG. 1--The mode-locked pulse width τ_p (FWHM) is shown as a function of the depth of modulation θ. The laser was Q-switched at a repetition rate in the range 75-300 Hz and was above threshold by a factor of 1.74. Losses include 18.6% output coupling and 7.4% internal losses.

This was experimentally verified by first tuning f_m to an acoustic resonance peak of the acousto-optic modulator and setting the cavity length to maximize the spectral width of the pulse. The laser was operating cw and the spectrum was monitored by a scanning Fabry-Perot interferometer. The gain used in (2) was the saturated gain g_0, which equals the sum of the cavity losses. The laser was then repetitively Q-switched at intervals much longer than the upper-state lifetime (240 μsec). The RF drive power to the acousto-optic Q-switch was set just below the level necessary for hold-off, thus ensuring that the gain during the period of cw oscillation and during the build-up interval was close to the unsaturated gain g, which was obtained from a knowledge of the flashlamp intensity in the pumping bands of Nd^{+3}:YAG as a function of current and from the measured threshold flashlamp current. The modulation frequency was then shifted the amount $\delta f_m'$ necessary to maximize the second harmonic power. From (2)

$$\delta f_m' = \frac{-2(g - g_0) f_m^2}{\pi \Delta f_a} \tag{3}$$

The experimental and theoretical frequency shifts agree quite well for the single data point taken.

$$(\delta f_m)_{exp} = -3.8 \text{ kHz} \tag{4a}$$

$$(\delta f_m)_{theory} = -4.1 \text{ kHz} \tag{4b}$$

The measurements shown in Fig. 1 were all obtained in this manner. Comparison of the plots of the intensity correlation function $G^{(2)}(\tau)$ obtained before and after peaking the second harmonic power demonstrates that the shortest pulses are obtained by maximizing the SH.

The transient mode-locking theory can be applied to other homogeneously broadened lasers which are Q-switched or pulsed, such as a low pressure CO_2 laser. The number of round trips required to attain steady-state mode-locked pulses is on the order of $\Delta f_a/\Delta f_{axial}$, which is typically between 10 and 100 for a CO_2 laser at one atmosphere pressure. Our results agree well with one published value of 1.24 nsec, for which the transient mode-locking theory predicts steady-state has been attained, making (1) applicable.

B. Experimental Studies of the Na-Xe Excimer Laser System

(T. L. Savarino and J. T. Yardley)

During the past three months, we have performed experimental work to complement the theoretical calculations done in the previous period. As described in the previous report, the object of this work is to arrive at a scheme for an efficient, tunable, visible laser. We have directed our attention to the Na-Xe molecule as a possible species for use in such a system. Specifically, we have performed measurements of fluorescent intensity versus wavelength, absorption versus wavelength, and various lifetimes associated with a suitably prepared sample which contained this molecule.

1. Absorption Measurements

Using a tungsten arc lamp (2200°K blackbody source) to illuminate it, we measured the overall absorption of a cell containing a few torr of Na vapor and 3 atmospheres of Xe, maintained at approximately 550°C. As a comparison, we also measured the absorption of a similarly prepared sample, substituting 3 atmospheres of He for the Xe. In both experiments and in all the work that followed at least 5 hours was allowed for the cells to reach equilibrium. Both experiments seemed to exhibit approximately similar absorption characteristics. In particular, we identified the following phenomena: (a) strong absorption of the Na D-line at 5890 Å; (b) regular undulations in the absorption spectrum with a spacing of 150 cm^{-1}, which we have attributed to the A band of diatomic Na; and (c) a strong absorption

doublet near 7690 Å, which we have attributed to Rb impurities in the Na. We are presently unable to discern whether the molecule Na-Xe is responsible for any of the absorption data taken. In the next period, we intend to measure the absorption of a cell which contains no foreign gas, in order to estimate the absorption due to diatomic Na alone.

2. Fluorescence Intensity

For this series of experiments, we measured the fluorescence of a cell prepared in the same manner as in the absorption measurements. We used a Nd:YAG laser with an output of 20 nsec pulses containing approximately 50 mJ of 1.06μ, which we frequency-doubled in a crystal of cesium dihydrogen arsenate and produced 1 mJ at 5320 Å. This was focused to a confocal parameter of 50 cm, centered in the cell. We have observed both a continuous and discrete line fluorescence spectrum in the region from 5300 Å to 10,000 Å using both an S1 and an S20 photocathode for sensitivity throughout the entire spectral region. A number of the discrete lines that have been observed have been ascribed to transitions between higher lying levels in the Na atom. We have formulated a hypothesis to describe why there is a significant number of Na atoms with electrons lying in states higher than $3p$. We believe that Na_2 molecules can collide with Na-Xe molecules producing dissociation, and leaving the Na atom in an excited electronic state. These excited Na atoms then absorb photons either from the pumping beam or from some other source inside the cell, which could be other relaxing Na atoms, Na_2 molecules, or Na-Xe molecules. This sequential absorption of photons by excited Na atoms could conceivably be quite common and lead to a large number of highly excited Na atoms.

We attempted to determine whether or not the observed fluorescence was strongly dependent upon the pressure of Xe in the cell. However, the present window seal construction will permit a maximum of 3 atmospheres of Xe. This is not enough dynamic pressure range to draw any valid conclusion about the pressure dependence of the fluorescence. We are in the process of constructing a cell which will withstand the higher pressures and high temperatures necessary to perform further measurements on this system.

We tried to determine the presence of, and measure, optical gain in the system. To do this, we aligned a He-Ne 6328 Å laser coincident with the pumping beam and looked for changes in the intensity at 6328 Å when the pump was on. We observed a pulse at 6328 Å which was 25% above the dc signal level. We measured its decay time to be approximately 1.5 μsec. However, because of the large measured loss at 6328 Å, we believe that this effect is due to a bleaching of the loss (probably due to Na_2) by the pumping radiation. The long decay time probably represents the Na_2 reformation time.

This result led us to conclude that the fluorescent intensity might depend very strongly on the intensity of the pumping radiation. We picked a wavelength region that we believed to be interesting (around 5900 Å) and observed that the fluorescence increased approximately as the pumping intensity to the third power. We believe that this indicates conclusively that we are bleaching the loss away in a very nonlinear fashion. Using the new cell we hope to repeat the experiment using higher Xe pressure and lower Na pressure in order to reduce the Na_2 concentration.

3. Lifetime Measurements

We attempted to measure the decay time of the fluorescent intensity and compare it to the decay time measured in the previous experiments. We measured a decay time of 300 nsec, five times less than the decay time measured in the gain experiments. Most of the measurements were made at 6160 Å for convenience. No strong wavelength dependence was observed. We are currently unable to explain this difference in a satisfactory manner. A possible explanation for the long lifetime could be in the argument previously given about sequential photon absorption to explain the higher lying electronic states of Na that we have detected.

In summary, during this period we have measured fluorescence intensity, loss, and fluorescent lifetime for the system of Na and Xe. In the next period, as we have indicated, we will use the high pressure, high temperature cell being constructed to extend these measurements to Xe pressure that we feel will be more favorable for formation of the molecule Na-Xe. According to the equilibrium constant and reaction rate which we have estimated, increasing the Xe pressure will move the equilibrium between Na-Xe molecules and isolated atoms closer to the molecules, thus increasing the fluorescence (and eventually we hope, increasing the possibility of optical gain).

C. Vacuum Ultraviolet Holography

(G. C. Bjorklund and S. E. Harris)

During this period we have continued to produce vacuum ultraviolet holograms using coherent 1182 Å radiation as the source of illumination. Holographic gratings with fringe spacings as fine as 836 Å were produced. Far-field Fraunhofer holograms of 1.3μ diameter particles were recorded on polymethyl methacrylate (PMM) and then read out by means of a scanning electron microscope.

The holographic gratings were, as before, produced by recording the linear fringe patterns which result from the interference between a plane object wave and a plane reference wave. The production of a holographic grating of 836 Å fringe spacing in PMM indicates that PMM has the resolution capability needed to record holograms produced by 1182 Å radiation with diffraction-limited resolution.

The far-field Fraunhofer method of holography is applicable to any object which consists of small, concentrated spots on a mostly transparent background. All that is required is to illuminate the object with a coherent wave and place the holographic recording medium in back of the object. A separate reference wave is not required. If the medium is separated from the object by at least one far-field distance (equal to several microns for typical objects) then the medium will record the interference between the unperturbed portion of the illuminating wave and the far-field pattern scattered by the object. This hologram could then be read out by

an electron microscope, expanded, and an enlarged image reconstructed using a visible wavelength laser to illuminate the expanded hologram. Alternatively, since the far-field pattern is merely the Fourier Transform of the object transmittance pattern, computer methods could be used to reconstruct the object image from the electron micrograph of the hologram.

The particles whose holograms were recorded were 1.305μ diameter latex spheres. These spheres are available in several carefully regulated sizes with diameters ranging from several microns down to 0.091μ. The spheres were supported on a 100 Å thick carbon film which was suspended approximately 20μ above the surface of the PMM. A carbon film of this thickness is about 30% transmitting to 1182 Å radiation. The carbon film was in turn supported by a 200 mesh grid. Each opening in the grid was individually marked by means of letter patterns in some of the grid bars. The PMM recorded both the holograms of the spheres and the shadow of the individually marked grid. Thus it was possible to positively locate the hologram of each particle on the carbon film. The holograms and the shadow of the grid were read out by means of a scanning electron microscope.

Work is presently in progress to record holograms of the smaller sized latex spheres and to utilize a transmission electron microscope for readout. Use of the transmission electron microscope offers an order of magnitude gain in resolution over that of the scanning electron microscope, as well as the possibility of a much more linear readout of the hologram fringe pattern. Once this linear readout is accomplished, computer techniques will be used to obtain reconstructed images of the spheres. A comparison of these reconstructed images with the known shape and size

of the spheres will indicate the actual resolution which may be achieved when this method of holographic microscopy is applied to biological subjects such as viruses and single cells.

D. Two-Photon Resonantly Pumped Frequency Converter

(S. E. Harris, J. F. Young, and D. M. Bloom)

In recent weeks we have developed the theory and performed first experiments on a new device which we term as a resonantly two-photon pumped frequency converter. This device is a further extension of our work on nonlinear optics in metallic vapors; and was also motivated by the four-frequency mixing technique demonstrated by Sorokin, Wynne, and Lankard.

A schematic of the basic device is shown in Fig. 1. The key idea is to utilize a tunable laser with an output frequency such that the sum of two of its photons equals a non-allowed transition of some metallic vapor. In Fig. 1 this driving frequency is termed as the pump laser and is at 4597 Å, so as to drive the non-allowed $3s$ to $4s$ transition in Mg. Physically, the effect of this pumping laser is to cause a pulsation or vibration of the electron cloud at twice the pump frequency. Since the transition is symmetric, there is no dipole moment, and thus no radiation, dispersion, or absorption. Any new frequency that is now incident on the Mg cell will now produce sidebands at $2\omega_p \pm \omega_t$, where ω_t is the frequency of this new, and rather arbitrarily tunable input frequency. We note immediately that this additional frequency need not be monochromatic and need not be supplied by a laser. The device may then function as a very efficient and versatile infrared up-converter and detector, and possibly infrared imaging device.

It is particularly important to note that the process described in the above paragraph is in general very broadband and that, for instance,

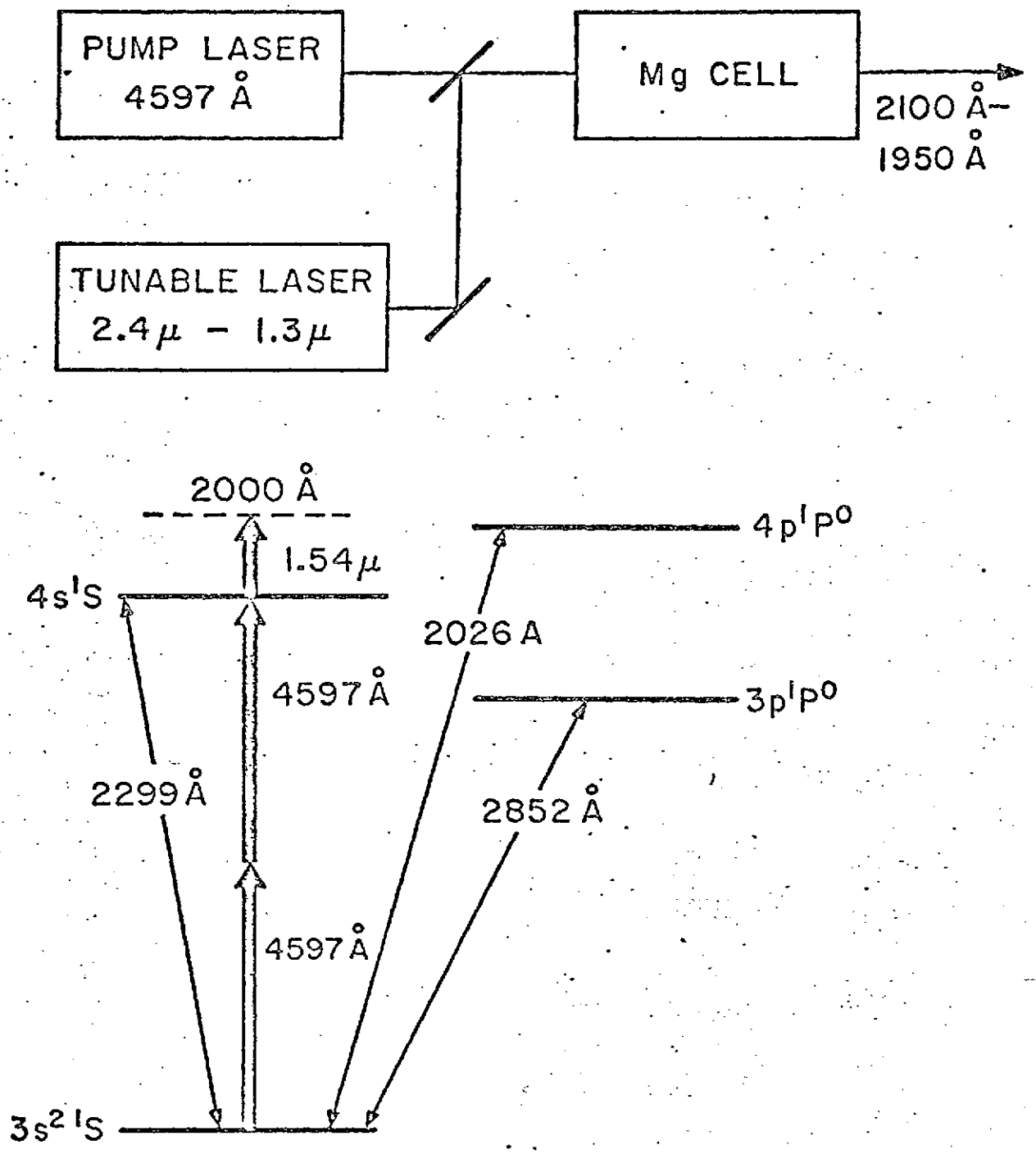

FIG. 1--Schematic of resonantly two-photon pumped frequency converter and pertinent energy levels of Mg .

it allows the infrared spectral region between perhaps 2μ and 20μ to be compressed and displayed into the middle of the visible spectrum. By making use of fast visible spectroscopic devices, for instance, the electronically tunable acousto-optic filter or fast scan vidicon devices, nearly the entire infrared spectrum can be instantaneously viewed.

In addition to its use as an infrared up-converter, the resonantly two-photon pumped frequency converter may be used to generate a very wide range of coherent radiation. Figure 1 shows its applicability for generation of tunable vacuum ultraviolet radiation. In this case, two photons of the pumping laser are mixed with a tunable photon which is variable over the range of 2.4μ to 1.3μ to yield an output tunable from 1950 Å to 2100 Å. Tunable infrared radiation may also be generated in this case by mixing two photons of the pumping frequency with a visible photon tunable in the vicinity of an upper p level.

The analysis of this device is given in the paper entitled "Resonantly Two-Photon Pumped Frequency Converter," which is included as Appendix A. This analysis proceeds by writing formulae for conversion efficiency, subject to the condition that the amplitude of the pumping frequency is limited by two-photon absorption of the non-allowed transition. We find the strikingly simple formula that conversion efficiency is given by

$$\text{Efficiency} = \frac{T_2}{\Delta t} \frac{\mu_{23}^2}{\mu_{03}^2}$$

In this equation T_2 is the dephasing time of the non-allowed transition, Δt is the length of the laser pulse, and μ_{23}^2 and μ_{03}^2 are matrix

elements connecting the respective levels, as described in Appendix A. For long-pulsed lasers Δt is replaced by T_1, the relaxation time of the upper level.

Note that the detuning from the non-allowed transition and thus implicitly the linewidth of the pump laser, does not enter into the conversion efficiency formula. Larger detunings require larger power densities but yield the same conversion efficiency. In general, conversion efficiencies predicted by this formula will vary between a few percent to in excess of 100% (only photon conversion efficiencies are limited to 100%). It is important to note that this formula predicts conversion efficiency in a single coherence length. If phase matching is used, conversion efficiencies may be further increased, although in this case certain of the advantages of this device (for instance, its very broad angular aperture, broad frequency bandwidth, and temperature insensitivity) may be sharply reduced. Further theory and details of the operation of this device are described in Appendix A.

A number of crucial experiments are necessary to ascertain the characteristics of this device. In a first experiment we have up-converted 1.06μ into the blue by the process $2\omega_p - \omega_t$, using a pumping photon at 6852 Å in Na vapor. Efficiencies in this first experiment are about 10^{-4}, which are close to what they should be at the conditions of this experiment. In a second experiment we have used the same two-photon pumped Na system to up-convert several lines of a CO_2 laser to the ultraviolet around 3320 Å. Photon conversion efficiencies in excess of 50% and power gains of about 12 dB were obtained. The experiment is described in detail in a paper

submitted to Applied Physics Letters and included as Appendix B. The experimental results of this experiment are also in good agreement with the theory.

With regard to conversion efficiency from infrared to visible, it should be noted that the NEP of a photomultiplier is about 7 orders of magnitude lower than that of a typical N_2 cooled infrared detector, and about 5 orders of magnitude lower than that of He cooled detectors. Thus, conversion efficiencies of between 10^{-5} to 10^{-7} of infrared power to visible power, break even with the present infrared detectors. Conversion efficiencies of this magnitude, or several orders of magnitude larger, may be obtained over reasonable areas and with large angular apertures. Since only one coherence length is employed, the angular apertures are limited only by geometrical considerations and not by phase matching. It is thus possible that practical imaging devices might be constructed.

As noted in Appendix A, the pumping laser is limited by two-photon absorption to a maximum power density of between 5×10^6 and 5×10^7 W/cm^2. The necessary pump power will thus be determined by the area of the focus of the pumping laser beam. This area is determined by the condition that the confocal parameter of the focus be approximately equal to one coherence length of the nonlinear specie. To the extent that the density of the nonlinear specie may be raised, the necessary pumping power is reduced. It is thus of practical importance to ascertain the upper limits of metal vapor pressure that may be employed. Rough estimates indicate that if pressures of perhaps 100 Torr of metal vapor can be used, that necessary pumping powers will be reduced to about 10 - 100 watts for conversion efficiencies in the 50% range.

A second key approach to increasing conversion efficiency will be to use a molecular quenching agent to reduce the lifetime of the excited non-allowed transition. Earlier workers have shown that 10 Torr of N_2 reduces the lifetime of allowed transitions to about 8 nsec.

Theoretical work on this project will be supported by the United States Army Research Office.

APPENDIX A

RESONANTLY TWO-PHOTON PUMPED FREQUENCY CONVERTER

by

S. E. Harris and D. M. Bloom

Preprint

M. L. Report No. 2232

November 1973

Contract No. DA-ARO-D-31-124-72-G-184

Contract No. NGL 05-020-103

Contract No. LASL #XP3-06745-1

Contract No. LLL 8532009

submitted to

Applied Physics Letters

Microwave Laboratory
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RESONANTLY TWO-PHOTON PUMPED FREQUENCY CONVERTER*

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(Received November 1973)

ABSTRACT

This letter describes a resonantly two-photon pumped frequency converter with application to the generation of tunable ultraviolet and vacuum ultraviolet radiation; and also to infrared to visible up-conversion and imaging. Calculations show that up-conversion power efficiencies in excess of 100% should be obtainable with tunable dye lasers having peak powers in the hundred watt to kW range.

*Work supported by the U.S. Army Research Office, the National Aeronautics and Space Administration, and the U.S. Atomic Energy Commission.

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In recent years, considerable success has been attained in utilizing metal vapors and mixtures of metal vapors and inert gases for frequency tripling of laser radiation into the ultraviolet and vacuum ultraviolet.[1,2] To date, this work has required picosecond time scale laser systems with peak powers in excess of 10^8 W . In this letter we describe a new technique which should allow generation of tunable UV and VUV radiation using any of a variety of tunable dye lasers and optical parametric oscillators having peak powers in excess of about 100 watts. The device described here may also act as a broadband IR to visible up-converter for coherent or incoherent IR radiation.

A schematic of the proposed device is shown in Fig. 1. To be specific, we consider the use of Mg to generate tunable UV and VUV radiation. A pump laser, in this case at 4597 Å , is tuned such that the sum of two photons is equal to the non-allowed $3s - 4s$ transition of Mg . A second tunable laser generates the sum or difference frequency, $2\omega_p \pm \omega_t$, where ω_p and ω_t are the frequencies of the pump and tunable lasers, respectively. This process will be particularly efficient if the generated frequency lies within a certain range of any of the $np\,^1P^0$ levels. For example, if the tunable laser is tuned over a range of about $\pm$ 1000 cm^{-1} centered at 1.7μ , then the

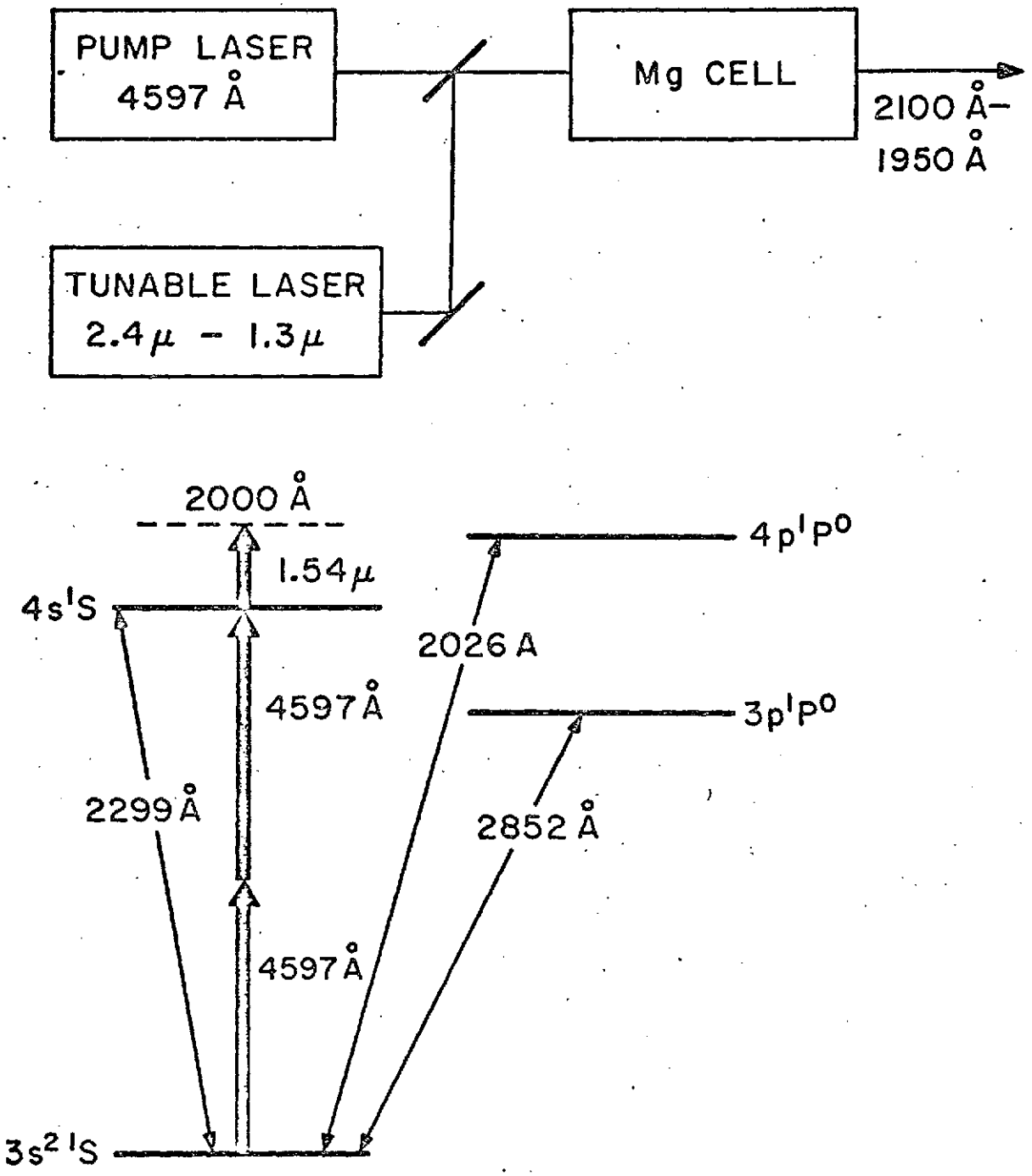

FIG. 1--Schematic of resonantly two-photon pumped frequency converter and pertinent energy levels of Mg .

theory to be derived below predicts approximately unity power conversion efficiency from tunable input to sum frequency centered at the $4p\,^1P^0$ level (2026 Å). A much broader range, about ± 10,000 cm^{-1}, centers at the $3p\,^1P^0$ level (2852 Å), though in this case the conversion efficiency will be several percent. For each higher np level, the conversion efficiency becomes higher, and the applicable range of this theory narrower.

As has long been known,[3] the advantages of using a non-allowed transition to resonantly enhance the nonlinear optical susceptibility are the absence of both loss and dispersion at the input and generated frequencies. However, as a result of the resonantly enhanced two-photon absorption, the power density of the pumping laser is restricted to a value which is several orders of magnitude lower than would be the case if this resonance had not been employed.

In the following, we write formulae for conversion efficiency from tunable laser power to sum (or difference) frequency power, with the assumption that the pump power density is limited by two-photon absorption of an intermediate non-allowed transition (for example, the $3s^2\,^1S - 4s\,^1S$ transition in Mg). The additional assumptions that the conversion occurs in a single coherence length, and that this length is determined strictly by the upper level (the $4p\,^1P^0$ level in Mg), lead to a strikingly simple formula for predicted conversion efficiency. Before proceeding, we note that resonant non-allowed transitions have recently been used in a somewhat different manner for the generation of tunable infrared radiation.[4]

From Maxwell's equations, the power density generated in a single coherence length at the sum frequency ω_s is given by $P/A\ (\omega_s) = (1/2\pi^2)\ \eta\omega_s^2 |\mathcal{P}(\omega_s)|^2\ L_c^2$, where $\mathcal{P}(\omega_s)$ is the generated dipole moment, L_c is the coherence length, and $\eta = (\mu/\epsilon_0)^{\frac{1}{2}}$. For an energy level system such as that shown in Fig. 1, the dipole moment and coherence length are closely approximated by

$$\mathcal{P}(\omega_s) = \frac{N\mu_{01}\mu_{12}\mu_{23}\mu_{30}\ E_p^2 E_t}{4\hbar^3\ (\Delta\omega_1)\left(\Delta\omega_2 + j\ \frac{\delta\omega_2}{2}\right)\Delta\omega_3} \tag{1}$$

and

$$L_c = \frac{2\pi\hbar}{N\eta}\ \frac{1}{\omega_s}\ \frac{\Delta\omega_3}{\mu_{03}^2}$$

where N is the atom density, μ_{ij} are the various dipole matrix elements, $\Delta\omega_1 = \omega_p - \omega_{01}$, $\Delta\omega_2 = 2\omega_p - \omega_{02}$, $\Delta\omega_3 = 2\omega_p \pm \omega_t - \omega_{03}$; where ω_p is the pump frequency, ω_t is the tunable frequency, and ω_{01} , ω_{02} , and ω_{03} denote the pertinent atomic transition frequencies from ground. (For the example of Fig. 1, these transitions occur to the $3p\,^1P^0$, $4s\,^1S$, and $4p\,^1P^0$ levels, respectively). $\delta\omega_2$ is the half-power linewidth of the ω_{02} transition, and E_p and E_t are the electric field strengths produced by the pump and tunable lasers. Combination of the above formulae leads to a predicted last coherence length conversion efficiency of

$$\mathcal{E} = \frac{\eta^2\mu_{01}^2\mu_{12}^2\mu_{23}^2}{\hbar^4\ \Delta\omega_1^2\ (\Delta\omega_2^2 + \delta\omega_2^2/4)\ \mu_{03}^2}\ (P/A)_p^2 \tag{2}$$

where $(P/A)_p$ is the power density of the pump laser.

We now determine the maximum allowed value of $(P/A)_p$ by the condition that $W^{(2)}\tau = \frac{1}{2}$, where $W^{(2)}$ is the two-photon transition probability and τ is the incident laser pulse length Δt or the decay time T_1 of the $4s^1S$ level, whichever is shorter. The maximum allowed power density of the pump laser is then[5]

$$(P/A)_{max} = \frac{\sqrt{2}\hbar^2 \Delta\omega_1}{\mu_{01}\mu_{12}\eta} \frac{1}{(\tau\delta\omega_2)^{\frac{1}{2}}} \left[(\Delta\omega_2)^2 + \left(\frac{\delta\omega_2}{2}\right)^2\right]^{\frac{1}{2}} \qquad (3)$$

We substitute this value of $(P/A)_{max}$ into Eq. (2) to obtain a conversion efficiency of

$$\varepsilon = \frac{T_2}{\Delta t}\frac{\mu_{23}^2}{\mu_{03}^2} = \frac{T_2}{T_1}\frac{\mu_{23}^2}{\mu_{03}^2} \qquad (4)$$

where T_2 [$T_2 = (2/\delta\omega_2)$] is the dephasing time of the $4s^1S$ level. The first and second equalities of Eq. (4) apply for laser pulse lengths less than T_1 and greater than T_1, respectively. (T_1 must include radiative trapping). For lasers with a pulse length longer than T_1, a molecular quencher may be used to substantially reduce T_1.[6] For the $4p^1P^0$ level of Mg, $\mu_{23}^2/\mu_{03}^2 = 48$.[7]

Note that the detuning from the $4s^1S$ level, and thus implicitly the linewidth of the pump laser, does not enter into the conversion efficiency formula. Larger detunings require larger power densities [Eq. (3)], but yield the same conversion efficiency. Before using Eq. (4), it must be ascertained that it is indeed the p level nearest the generated signal

that determines the coherence length. An approximate condition for the validity of this assumption is that $\mu_{03}^2/\Delta\omega_3 > \mu_{01}^2/\Delta\omega_1$. We should note that if phase matching techniques[1] are used to further increase the predicted conversion efficiency, that the increase will only be linear with the number of coherence lengths which are matched. This results since increasing the number of coherence lengths reduces the allowable value of $W^{(2)}\tau$ and thus of $(P/A)_{max}$.

As a first example, consider a Mg cell at a vapor pressure of 10 Torr. Assuming a laser pulse length of 4 nsec and a $4s^1S$ linewidth of 0.1 cm^{-1} , then for conversion to the vicinity of the $4p^1P^0$ level, Eq. (4) yields an efficiency of 126% . (Note that only photon conversion efficiencies are limited to 100%). Assuming $\Delta\omega_2 = 0.1\ cm^{-1}$, we require a pump power density of 9.7×10^7 watts/cm^2 . For this case, the range of validity of Eq. (4) is about $\pm$ 1000 cm^{-1} . At the ends of this band, at a pressure of 10 Torr, the coherence length will be 0.5 cm long; and thus for confocal focusing, a pumping laser power of 557 watts is required.

As noted earlier, other applicable ranges of this theory center at each of the np^1P^0 levels. Table I gives a number of other examples of metal vapor systems and applicable ranges for the generation of tunable UV and VUV radiation.

As another and somewhat different example, we consider the use of resonantly two-photon pumped Na for conversion of an IR signal at 10.6μ into the near ultraviolet. For a pump laser at 6856 Å (i.e., two-photon pumping of the $3s - 3d$ transition), a 10.6μ signal will be converted to 3320 Å . Since in this case the generated frequency is somewhat outside

TABLE I

SYSTEMS FOR ULTRAVIOLET AND VACUUM ULTRAVIOLET GENERATION

ELEMENT	PUMP WAVELENGTH	TUNABLE WAVELENGTH	GENERATED WAVELENGTH
Mg	4s: 4597 Å 3d: 4310 Å	2.4μ → 1.3μ (-) 0.467μ → 11.0μ	2100 Å → 1950 Å 4000 Å → 2200 Å
Cd	6s: 3752 Å 5d: 3377 Å	1.8μ → 1.4μ (-) 0.395μ → 1.9μ	1700 Å → 1650 Å 2950 Å → 1850 Å
Zn	5s: 3585 Å 4d: 3202 Å	1.5μ → 1.1μ (-) 0.393μ → 1.9μ	1600 Å → 1550 Å 2700 Å → 1750 Å
Hg	7s: 3129 Å 6d: 2804 Å	1.6μ → 1.1μ (-) 0.372μ → 1.5μ	1425 Å → 1375 Å 2250 Å → 1550 Å

The (-) signs denote the difference frequency process $\omega_s = 2\omega_p - \omega_t$.

the allowed range of this theory, a more exact computer-calculation was employed. Assuming $\Delta\omega_2 = \delta\omega_2 = 0.1\ cm^{-1}$ and Δt or $T_1 = 10$ nsec, we find a conversion efficiency of 8.8% at an allowed incident power density of $5.25 \times 10^6\ W/cm^2$. The conversion will be very broadband, allowing up-conversion and imaging of thermal radiation.

The authors acknowledge a stimulating discussion with R. Hodgson, who has also been interested in devices of this type. Helpful discussions with A. H. Kung, E. A. Stappaerts, and J. F. Young are acknowledged.

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APPENDIX B

INFRARED UP-CONVERSION WITH RESONANTLY TWO-PHOTON PUMPED METAL VAPORS

by

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Preprint

M. L. Report No. 2245

December 1973

Contract Nos.

F19628-73-C-0025

NGL 05-020-103

Navy 44

submitted to

Applied Physics Letters

Microwave Laboratory
W. W. Hansen Laboratories of Physics
Stanford University
Stanford, California

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by

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(Received December 1973)

ABSTRACT

We report efficient up-conversion of low-level IR radiation near 10μ to the near ultraviolet. Radiation at 9.26μ is converted to 3505 Å with a photon conversion efficiency of 58% and a corresponding power gain of 16.2. The process employs resonant two-photon pumping of the non-allowed 3s - 3d transition in Na.

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*Work supported by the Air Force Cambridge Research Laboratories, the National Aeronautics and Space Administration, and by the Joint Services Electronics Program through the Office of Naval Research.

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INFRARED UP-CONVERSION WITH RESONANTLY TWO-PHOTON PUMPED METAL VAPORS

by

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We report efficient up-conversion of IR radiation at 10.61μ, 10.23μ, 9.57μ, and 9.26μ to the near ultraviolet at 3321 Å, 3317 Å, 3309 Å, and 3305 Å, respectively. The process makes use of a resonantly enhanced third-order nonlinear susceptibility, which is achieved by two-photon pumping of the non-allowed $3s - 3d$ transition of Na.[1-4] A photon conversion efficiency of 58% and a corresponding power gain of 16.2 is obtained for the 9.26μ conversion. These conversion efficiencies are obtained in a single coherence length of metal vapor, and are therefore quite broadband in both wavelength and angular aperture.

A schematic of the resonant up-converter is shown in Fig. 1. Pumping radiation at $\omega_p = 6856$ Å, obtained from a pulsed Nd:YAG pumped $LiNbO_3$ optical parametric oscillator, and IR radiation obtained from a cw CO_2 laser are incident on a Na cell at a vapor pressure of about 1 Torr. The optical parametric oscillator (OPO) is tuned to resonantly excite the non-allowed $3s - 3d$ Na transition at 3428 Å. Although strongly excited, this transition has no dipole moment and does not radiate. Incident radiation

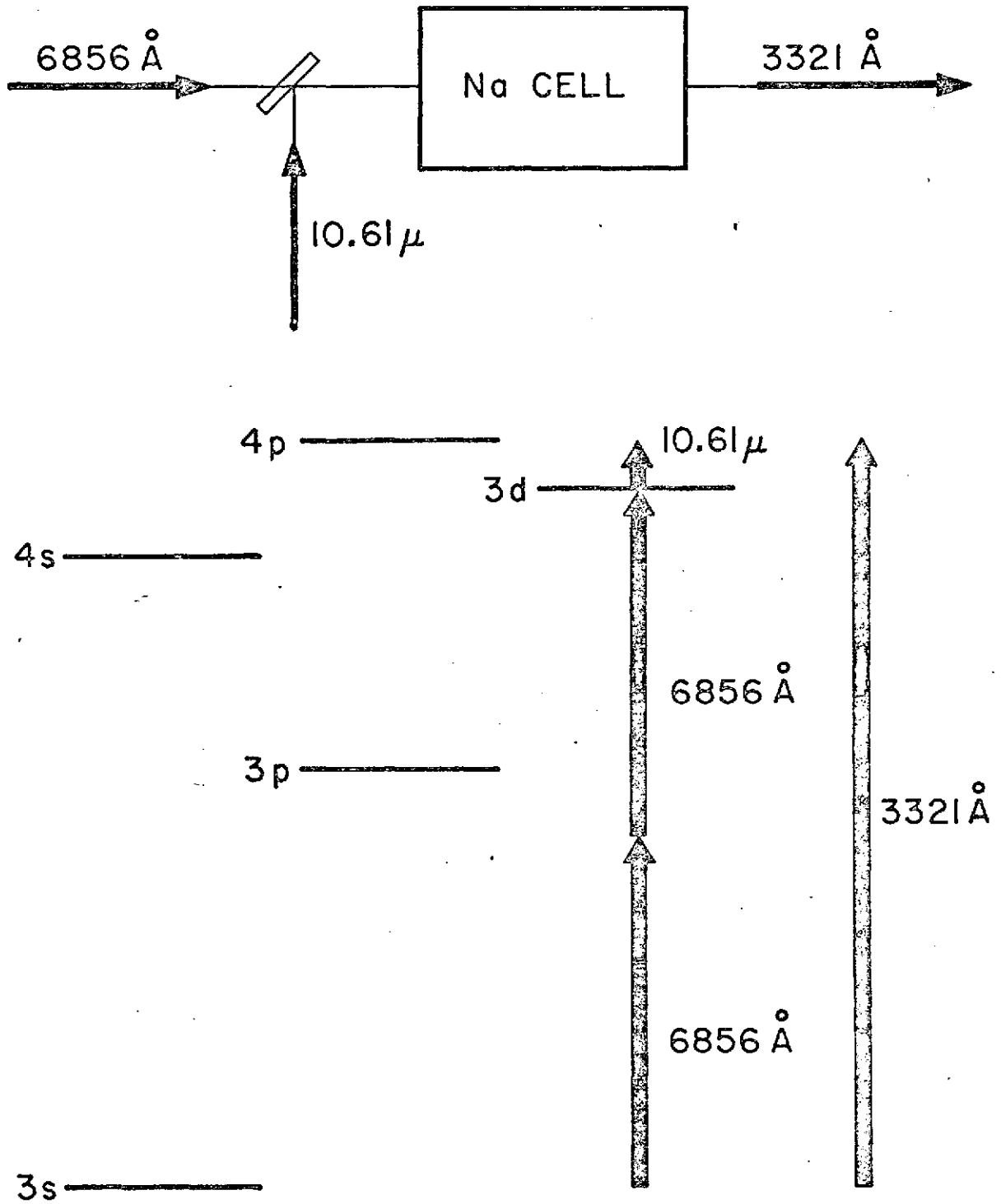

FIG. 1--Schematic of IR frequency conversion experiment.

at frequency ω_t couples this symmetric excitation to an allowed transition to ground, and produces radiation at $2\omega_p \pm \omega_t$. The upper sideband $(2\omega_p + \omega_t)$ is much closer to an allowed p-ground transition, and is therefore several orders of magnitude stronger than is the lower sideband.

The advantage of using a non-allowed transition to resonantly enhance the nonlinear optical susceptibility is the absence of both loss and dispersion at both the input and generated frequencies.[5] However, as a result of increased two-photon absorption, the power density of the pumping laser is restricted to a value several orders of magnitude lower than would be the case if this resonance had not been employed. The theory of this process is examined by Harris and Bloom,[1] who show that power conversion efficiencies in excess of 100% should be obtainable in a single coherence length of metal vapor. We note that resonant non-allowed transitions have recently been used in a somewhat different manner for the generation of tunable infrared radiation.[4]

In the present experiment, the OPO had a peak output power of 3 kW, a pulse length of 20 nsec, and a linewidth of 2 cm^{-1}. It was focused into the Na cell to a confocal parameter of 2 cm, with a power density of about 10 MW/cm^2 at the beam waist. The CO_2 laser was incident to the cell at a cw power of about 5 mw. The CO_2 laser was tuned between various output frequencies by adding a small amount of SF_6. The intensity of radiation at the UV sum frequency was measured with a carefully calibrated Spex monochromator and photomultiplier. Measured power conversion efficiencies from infrared to ultraviolet were as follows: $10.6\mu \rightarrow 3321$ Å, 7.9% ; $10.23\mu \rightarrow 3317$ Å, 10.9% ; $9.57\mu \rightarrow 3309$ Å, 108% ; and $9.26\mu \rightarrow 3305$ Å,

1620%. These power efficiencies correspond to photon efficiencies of 0.25%, 0.35%, 3.7%, and 58%, respectively. Once a Na pressure corresponding to one coherence length was reached (about 1 Torr), conversion efficiencies were quite insensitive to Na pressure. The sharply increasing conversion efficiency as the input frequency is varied from 10.61μ to 9.26μ results from the approach of the sum frequency to the $3s - 4p$ transition frequency of Na. Conversion efficiencies are in close agreement with the theory of Reference 1.

Two other interesting effects were observed during the course of this experiment. The first was the observation of a four-frequency parametric oscillation, with an output frequency greater than the 6856 Å pump. As the OPO was tuned to the $3s - 3d$ two-photon transition, a polarized, somewhat conical emission centered at a signal frequency of 5904 Å was observed. A search revealed the presence of the idler frequency at 8173 Å; i.e., such that signal plus idler frequency equal twice the pump frequency. The second interesting observation was a weak, polarized, collimated emission at twice the 6856 Å pump frequency.

We should note that although the pumping power density in this experiment was 10 MW/cm^2, theory[1] predicts that for constant conversion efficiency, this power density may be reduced linearly with the linewidth of the pumping laser. We thus expect that at a pump linewidth of 0.1 cm^{-1} that comparable conversion efficiencies should be obtainable at a pumping density of about 5×10^5 $watts/cm^2$. By increasing the Na pressure to reduce the coherence length to several mm, it may be possible to obtain these conversion efficiencies at powers of 10 or 100 watts. By operating at reduced efficiencies,

large areas and angular apertures should be obtainable. We also note that the non-allowed transition may be excited by any combination of frequencies. For instance, Nd:YAG radiation at 1.064μ and R6G radiation at 6119 Å may be used to excite the $3s - 4s$ Na transition.

The device described here has a number of potentially important advantages for IR up-conversion and imaging.[6-9] These include angular aperture, bandwidth, and the fact that the generated frequency $(2\omega_p + \omega_t)$ is far removed from the pump frequency ω_p, and lies in a region of excellent photocathodes. A disadvantage, as compared to nonlinear crystal up-conversion is the power square dependence of conversion efficiency.

This device may also be used as a generator for UV, VUV, and IR radiation.[1,10] For instance, if radiation in the vicinity of 3310 Å is incident on 6856 Å two-photon pumped Na, then high quantum efficiency for generation in the 9.0μ – 10μ region should be obtainable. Another interesting experiment would be to use a higher power CO_2 source (about several hundred watts) to deplete the red pump at 6856 Å. This technique should allow efficient conversion of light from flashlamp pumped dye lasers into the UV and VUV. Phasematching techniques might also be combined with the two-photon pumping technique described here.[1-3]

The authors acknowledge helpful discussions with R. L. Byer, R. L. Herbst, A. H. Kung, and A. E. Siegman.

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